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Thin Solid Films

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## ABSTRACT

Lasers for fusion experiments use thin-film dielectric coatings for reflecting, antireflecting and polarizing surface elements. The most important requirements of these coatings are uniformity, accuracy and laser damage threshold. Among the current fusion lasers, carbon dioxide, iodine and Nd:glass, coatings are most important to the Nd:glass laser. There, damage resistance in particular strongly affects the laser's design, performance and operating cost. The success of advanced lasers for future experiments and potential reactor applications will require significant developments in damage resistant coatings for ultraviolet laser radiation.

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# Optical Coatings for Laser Fusion Applications

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## 1. Introduction

Experiments in inertial confinement fusion are being conducted at laboratories throughout the world. The goal of these programs is to heat and compress a DT fuel mixture to one-hundred-million degrees Centigrade and one-thousand times liquid density. At such high temperature and density, thermonuclear burning of the fuel occurs with subsequent release of energy. The success of inertial confinement fusion rests on our ability to deliver sufficient energy and power to the fuel in a properly shaped pulse. Meaningful experiments now require delivering 10-100 kJ of energy at power levels of 10-100 TW, and the energy and power required for eventual reactor applications may be ten times greater.

The methods currently receiving greatest attention for delivering such enormous energy and power to a submillimeter gas-filled target are lasers and particle beams of electrons, light ions and heavy ions. Of these potential sources, lasers are the most well-developed and widely applied for fusion experiments. The lasers currently used are (1) carbon dioxide with a wavelength of 10.6 $\mu$m, (2) atomic iodine at 1.32 $\mu$m, and (3) glass doped with neodymium ions which lase at 1.06 $\mu$m. Recently, several laboratories have begun fusion experiments using the second, third and fourth harmonic frequencies of Nd:glass radiation with wavelengths of 0.53 μm, 0.35 μm, and 0.27 μm. In addition, effort to develop the 0.248-μm wavelength KrF laser for fusion studies has recently been accelerated.

Thin film coatings are used in all these laser systems. The importance of coatings to overall performance of the system varies greatly; having presently the greatest impact on Nd:glass lasers. For example, "Shiva" the 20 beam laser at Lawrence Livermore Laboratory, has 2500 optical elements, of which 2000 are coated with dielectric thin films.

Following this introduction, Section 2 reviews the optical coating applications for each fusion laser system. Laser design issues related to coatings are presented in Section 3. Section 4 discusses the importance of laser-induced damage to coatings and status of damage experiments. Promising areas for future development are reviewed in Section 5.

## 2. Coatings in Fusion Lasers

Thin-film coatings have three optical applications in fusion lasers: (1) high-reflection (HR) coatings for mirror surfaces, (2) anti-reflection (AR) coatings on the surfaces of lenses and windows, and (3) polarizing beamsplitters used to control the direction of beam propagation. Designs for these multilayer coatings are reviewed elsewhere in these proceedings.[1]

### 2.1 $CO_2$ Laser

The largest operating $CO_2$ laser for fusion studies is the eight beam "Helios" at the Los Alamos Scientific Laboratory. Each

40-cm-aperture beam of Helios can generate more than 1000 J in pulsewidths less than 1 ns; giving the total laser an output capability greater than 8 kJ at 8 TW. The 72-beam "Antares" now under construction at LASL, is expected to produce 100 kJ at 100 TW when completed.

The following coatings are used in $CO_2$ lasers:

- AR - NaF on NaCl substrates for target chamber and amplifier windows, ZnS single layer or $ZnS/ThF_4$ multilayer on Ge substrates for output coupler and modelocker.
- HR - $ZnS/ThF_4$ on aluminum coated copper.
- Polarizer - $ZnS/ThF_4$ or a Au grid, both deposited on ZnSe substrates.

The most important coating is the NaF AR coating on NaCl target chamber and amplifier windows. This coating has a damage threshold of 6 $J/cm^2$ for 1-ns, 10.6-$\mu$m pulses, which substantially exceeds the typical operating fluence of 1 $J/cm^2$, and equals the bare-surface threshold of optically polished NaCl. The $ZnS/ThF_4$ polarizer has 1-ns damage thresholds of 2 $J/cm^2$ and 9 $J/cm^2$ for 10.6- m light with p and s polarization respectively.[2]

## 2.2 Iodine Laser

The largest iodine laser for fusion studies is the single beam Asterix III at the Max Planck Institute in Garching, West Germany. This laser has produced 300-J, 250-ps, 1.2-TW pulses from a 17-cm diameter aperture[3] with a fluence loading of 2 $J/cm^2$ on the output amplifier window and focusing optics. Multilayer AR, HR and polarizing coatings of $SiO_2/TiO_2$ are used. Coating applications in the iodine laser are similar to those in the Nd:glass laser. However, amplifier staging of Asterix III was not optimized to take full advantage of coating damage resistance. Consequently coatings do not currently limit its performance.

## 2.3 Nd:Glass Laser

The 20 beam Shiva at Lawrence Livermore Laboratory is the world's largest operational Nd:glass laser. Each 20-cm-aperture beam can produce 750-J, 1-ns pulses. The full laser generated a record power of 27 TW at shorter pulsewidth. Nova, scheduled for operation in 1983, will produce 100 kJ at 100 TW power in 1-ns pulses from 10 beams, each of 74-cm aperture. Multilayer $SiO_2/TiO_2$ AR, HR and polarizing coatings are used throughout the lasers. In fact, all optical element surfaces, except the glass amplifier disks have dielectric coatings. In contrast to the $CO_2$ and iodine lasers, whose power and energy is limited in the current designs by gain saturation, the Nd:glass laser's performance is limited by laser-induced damage to these coated surfaces.

## 2.4 KrF Laser

A KrF fusion laser module is being developed to produce 10 kJ in 10-ns pulses giving a 1-TW output power at peak fluence loading of 5 $J/cm^2$. Achieving adequate damage thresholds for thin-film AR and HR coatings is a key element for successful development of the KrF laser.

# 3. Design Requirements

The central importance of thin-film coatings in high energy, Nd:glass "hot spots" from the beam, caused by the intensity-dependent refractive index of glass, and (3) isolation stages, consisting of Faraday rotators between crossed thin-film polarizers, to allow light to pass only in the forward direction, toward the target. As the pulse energy increases, its diameter and the aperture of these elements are expanded to maintain constant fluence loading. Turning mirrors reflect the high energy pulse to the evacuated target chamber, where it passes through a window, low f-number lens, and a thin glass plate, which shields the lens from damage by target debris.

Except for the amplifier disks, all optical surfaces are coated with dielectric thin films. There are AR coatings on spatial-filter lenses, Faraday-rotator glass, target-chamber windows, focus lenses and debris shields. Other surfaces have polarizing or HR coatings deposited on BK-7 glass substrates. Reflecting and antireflecting coatings also are used on elements in the beam diagnostic packages.

The most important requirements of optical coatings for fusion laser applications are:

- Uniformity
- Accuracy
- Damage threshold .

"Accuracy" represents the average value of reflectance and transmittance over the surface, while "uniformity" refers to local variations from the average. Uniformity is normally more important than accuracy because, while small variations from the average can be compensated by adjusting amplifier gain, lack of uniformity usually results from variations in layer thickness and causes a wavefront error in the beam in addition to the amplitude variation. Wavefront errors affect beam propagation and focusing onto the target. The wavefront error of transmitting and reflecting elements which can be allowed is one-tenth wave for HeNe laser light (632.8-nm wavelength). A summary of the design specifications for Shiva coatings is given in Table 1.

Table 1

Specifications for Shiva Coatings

| Coating | Accuracy | Uniformity |
|---|---|---|
| Mirror | $R \geq 99\%$ | $\pm$ 0.2% |
| Beamsplitter | $R \leq 90\%$ ($\pm$ 1.5%) | $\pm$ 0.1% |
| Polarizer | R (s pol.) $\geq$ 98.5% | $\pm$ 0.2% |
| | R (p pol.) $\leq$ 3.0% | $\pm$ 0.3% |
| Antireflector | $R \leq 0.2\%$ | --- |

Maximum apertures of coated elements for Nova will be 80 cm for AR coatings, 72 cm for polarizers and 109 cm for HR coatings. Polarizers and beamsplitters present the greatest production difficulties because of their large number of layers and sensitivity of the coating's performance to errors in layer thickness.

"Damage threshold" is the fluence (energy per unit area in the laser pulse) which begins to cause irreversible physical change in the coating. Nd:glass lasers are designed to operate at fluence levels just below the damage threshold. Thus, we require the greatest possible damage resistance to minimize the laser system's aperture and thus its cost. This requirement translates in practice to exercising extreme care in preparing and cleaning substrate surfaces, eliminating spatter and other coating defects, and maintaining correct stoichiometry on a microscopic scale.

It is important also that coating properties do not change after a period of time. We are concerned in particular with possible spectral shifts of polarizers and with changes in damage fluence. We have observed, for example, the damage threshold of some HR coatings to decrease by one-half after storage for a year in a laboratory environment.

Optical elements of lasers normally are handled with great care in environments in which dust, humidity and temperature are controlled so physical durability, abrasion resistance and adherence are less important than in other applications.

## 4. Laser Damage to Coatings

Because laser-induced damage to coatings is very important in the design and performance of Nd:glass fusion lasers, we have devoted great effort to understanding the causes of laser damage and to developing materials and deposition processes which improve damage thresholds.[4]

Laser damage is caused by absorption of light in the coating. The absorbed energy increases the temperature in a small volume, leading to thermal-stress fracture or melting. The major sources of absorption in transparent dielectrics are (1) impurities, defects or deviations in stoichiometry and (2) plasmas generated by electron-avalanche ionization. Importance of the avalanche-ionization mechanism is greatest for pulses of subnanosecond duration. Linear absorption by impurities dominates the damage process for nanosecond-and-longer pulse widths, which is the regime of greatest interest for laser fusion experiments. Laser calorimetry[5] has been recently developed which allows measurement of linear absorption with sensitivity of 1 part in $10^5$. The absorption coefficients measured for thin films lie typically in the range $1\text{-}10^3$ $cm^{-1}$. For comparison, optical glass has absorption coefficients of $10^{-4}$ - $10^{-3}$ $cm^{-1}$, and in the interface region between film and substrate we estimate coefficients to be in the range of $10^2$ - $10^4$ $cm^{-1}$. These absorption coefficients are the average value over the volume of the coating which is sampled by the laser beam. We expect the absorption at localized impurity sites to be much greater.

## 4.1 AR Coatings

The AR coating on input spatial-filter lenses is the most vulnerable to damage of all coatings in the laser system. This coating receives the greatest fluence loading, and generally AR coatings have lower damage thresholds than other coatings. The major cause of low AR thresholds is, we believe, that the substrate interface region, with its large absorption coefficient, is exposed to the electric field of the laser pulse in AR, but not in HR coatings. The interface of polarizer coatings also is exposed to the field of p-polarized light, but fluence loading is reduced by the geometrical factor associated with use of the coatings at Brewster's angle. The evolution of damage morphology for an AR coating, which is shown by the electron-beam microscope photographs in Fig. 2,[6] gives further evidence that AR damage begins at the substrate interface.

Our attempts to improve damage thresholds of AR coatings have therefore emphasized surface preparation as well as substrate and coating materials. Fig. 3 shows a comparison of damage thresholds measured for 1-ns pulses of 1.064-$\mu$m light on $SiO_2/TiO_2$ AR coatings which are deposited on both conventional-and bowl-feed-polished fused-silica substrates.[7] AR coatings on bowl-feed polished substrates have greater damage thresholds for reasons which are probably associated with the chemical composition or "cleanability" of the smoother bowl-feed polished surface. We found no difference in thresholds of coatings deposited on fused silica and those deposited on the standard optical glass, BK-7. However, a half-wave-thick silica "undercoat" layer deposited on either of the two substrate materials before the AR coating increased the median damage threshold by 30%.

We studied damage thresholds of AR coatings made of many different materials, all deposited by electron-beam evaporation. Among oxide coatings we tried $SiO_2$ in combination with one of the higher-index materials $TiO_2$, $Ta_2O_5$, $ZrO_2$ and $Al_2O_3$. We have also examined the fluoride coatings $MgF_2$, NaF, $Na_3AlF_6$, $MgF_2/ThF_4$, $MgF_2/PbF_2$, $ZnS/ThF_4$ and $MgF_2$ with an $Al_2O_3$ overcoat. Although we occassionally found some outstanding examples among these coatings, none of them surpassed the average performance of our standard $SiO_2/TiO_2$ coating.

We then systemmatically studied the influence of the major deposition variables: temperature, rate and oxygen pressure on damage thresholds of $SiO_2/TiO_2$ and $SiO_2/Ta_2O_5$ coatings. The results indicate that lower temperature coatings have improved thresholds.

In other studies we found damage thresholds increased for coatings with smaller gain size,[8] and a somewhat surprising lack of correlation between damage threshold and each of the properties: absorption, stress and adhesion.[9] We believe the reason for this lack of correlation is these are macroscopic average properties, while damage is caused by the microscopic localized features of the coating.

4.2 HR Coatings

Damage thresholds of HR coatings do not depend on substrate preparation or interface condition because the electric field amplitude of the laser pulse decreases rapidly with distance into the coating. Damage therefore typically occurs in the outer layers of HR coatings; frequently at defect or impurity sites which can be seen with a microscope before laser irradiation.

Our experiments with HR coatings have emphasized increasing mechanical strength and reducing the electric field strength at layer interfaces. The most significant improvement was to add a half-wave-thick silica "overcoat" to the normal quarter-wave HR stack. Overcoats have been used to improve durability and abrasion resistance. Their effect on damage threshold is shown by the two histograms in Fig. 4. A likely explanation for the increased thresholds of overcoated mirrors is that the thick, amorphous silica layer, under compressive stress, inhibits rupture of the underlying, microcrystalline $TiO_2$ layer, which is in tension.

We studied the relation between average absorption and damage threshold for a series of HR coatings prepared by OCLI. The coatings had absorptions ranging from $10^{-2}$ to $10^{-5}$, which depended on the oxygen pressure during deposition. Damage thresholds generally decreased with increasing absorption for coatings with absorption above $10^{-2}$. However, among coatings whose absorptions were less than $10^{-2}$, threshold was independent of the average absorption. This result was not unexpected because damage results from absorption by localized impurities, which can greatly exceed the average value.

In a second experiment, OCLI prepared a series of HR coatings whose designs were altered from the normal quarter-wave stack so as to reduce the peak or average electric field intensity in the outermost $TiO_2$ layer or at the first $TiO_2/SiO_2$ interface. These modified-field designs were expected to reduce the total absorption, which we believe is concentrated at interfaces and in the high index layers. However, in these two cases we found respectively no change in threshold and the opposite change expected. Variations in coating stress caused by different layer thicknesses may have been responsible for these unexpected results, although it is more likely that the dependence of damage on local defects was stronger than changes in the average field strength and absorption.

## 5. Areas for future coating development

Areas for future development which will significantly impact the design, performance and cost of fusion lasers are:

- Special application coatings
- Alternate deposition technologies
- Coatings for UV application

Examples of special application coatings which are currently under development are:

(1) Durable coatings which can be chemically stripped without damage to the substrate surface. Use of these coatings would reduce operating costs by eliminating expensive refinishing of the optical surfaces before recoating the damaged element. Our studies of strippable AR coatings which have a cryolite layer next to the substrate have given promising results.[10]

(2) Durable coatings deposited at room temperature. Such coatings are required for temperature sensitive glass and crystals.

(3) Transparent conductive coatings with damage threshold comparable to AR coatings.[11] These coatings may permit fabrication of large aperture electro-optic switches and possibly cause dramatic changes in the basic architecture of fusion lasers.

There are deposition technologies not ordinarily used for optical coatings that have the potential to produce damage resistant coatings. Among the possibilities are: (1) oxide coatings deposited from metal-organic solutions,[12] (2) chemical vapor deposition, (3) deposition in ultra-high vacuum, and (4) viscous liquid coatings which flow continuously over the element surface. Improvements may also come from preparing substrate surfaces in the evacuated coating chamber by such methods as: (1) surface etching with laser, electron or ion beams, (2) high temperature baking, or (3) strong UV irradiation.

Finally, the development of damage resistant coatings for UV fusion laser applications requires immediate attention. The coatings should withstand 5 J/cm$^2$ fluence of 0.25-$\mu$m wavelength radiation, and must survive in the corrosive, fluorine gas environment, exposed to the effects of a high-voltage electric discharge, including energetic electrons and vacuum-UV radiation. Successful coatings will probably be high-band-gap oxides and fluorides deposited with great attention to purity and cleanliness.

In conclusion, thin film coatings play a central role in the design, performance, and cost of lasers for fusion experiments. Recent, substantial improvements in coating damage thresholds were required for the Nd:glass laser, Nova, which will be the primary laser for fusion studies in the mid-1980's. Development of UV lasers for fusion experiments and possible reactor applications depends critically on improvements in damage resistant UV coatings.

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3596P

## References


1. J. H. Apfel, these proceedings, paper number RE 13.
2. C. R. Phipps, Jr., Los Alamos Scientific Laboratory, private conversation.
3. G. Brederlow, R. Brodman, K. Eidmann, M. Nippus, R. Petsch, S. Witkowski, R. Volk, and K. H. Witte, IEEE J. Quant. Electr., QE-16, 122 (1980).
4. W. H. Lowdermilk, D. Milam, and F. Rainer, in Laser Induced Damage in Optical Materials: 1979, NBS Special Publication, A. Glass, A. Guenther, H. Bennett and B. Newnam, eds. (1979).
5. D. L. Decker and P. A. Temple in Laser Induced Damage in Optical Materials: 1977, NBS Special Publication 509, p. 281 (1978); T. H. Allen, J. H. Apfel and C. K. Carniglia in Laser Induced Damage in Optical Materials: 1978, NBS Special Publication 541, p. 33 (1979).
6. Electron-beam microscopy by G. B. Carrier of Corning Glass Works, Corning, N. Y.
7. These and other coatings, except those specifically identified otherwise, were prepared by Optical Coating Laboratory, Inc.
8. Coatings were prepared by Pacific Northwest Laboratories.
9. These experiments were performed in collaboration with C. K. Carniglia, T. H. Allen and T. A. Tuttle of OCLI and Y. Nakajima of Hoya Corporation.
10. G. R. Wirtenson, Lawrence Livermore Laboratory, private conversation.
11. W. T. Pawlewicz, I. B. Mann, W. H. Lowdermilk and D. Milam, Appl. Phys. Lett. 34, 196 (1979).
12. H. Schroeder in Physics of Thin Films, Advances in Research and Development, Vol. 5, G. Hass and R. E. Thun, eds. (Academic Press, New York, 1969).

## Figure Captions

Figure 1. Schematic diagram showing components in one beam of the Nd:glass laser Nova, under construction at LLL.

Figure 2. Morphology of damage to AR coating, photographed using electron-beam microscope.[6] Laser pulse fluence increased from (a) to (d). Width of each photographed region was 3 $\mu$m.

Figure 3. Comparison of 1-ns 1.06-$\mu$m pulse damage thresholds of $SiO_2/TiO_2$ AR coatings deposited on (a) conventionally polished surface and (b) bowl-feed polished surface.

Figure 4. Comparison of 1-ns, 1.06-$\mu$m pulse damage thresholds of $SiO_2/TiO_2$ HR coatings: (a) normal quarter-wave stack without overcoat, (b) quarter-wave stack with half-wave-thick silica overcoat.

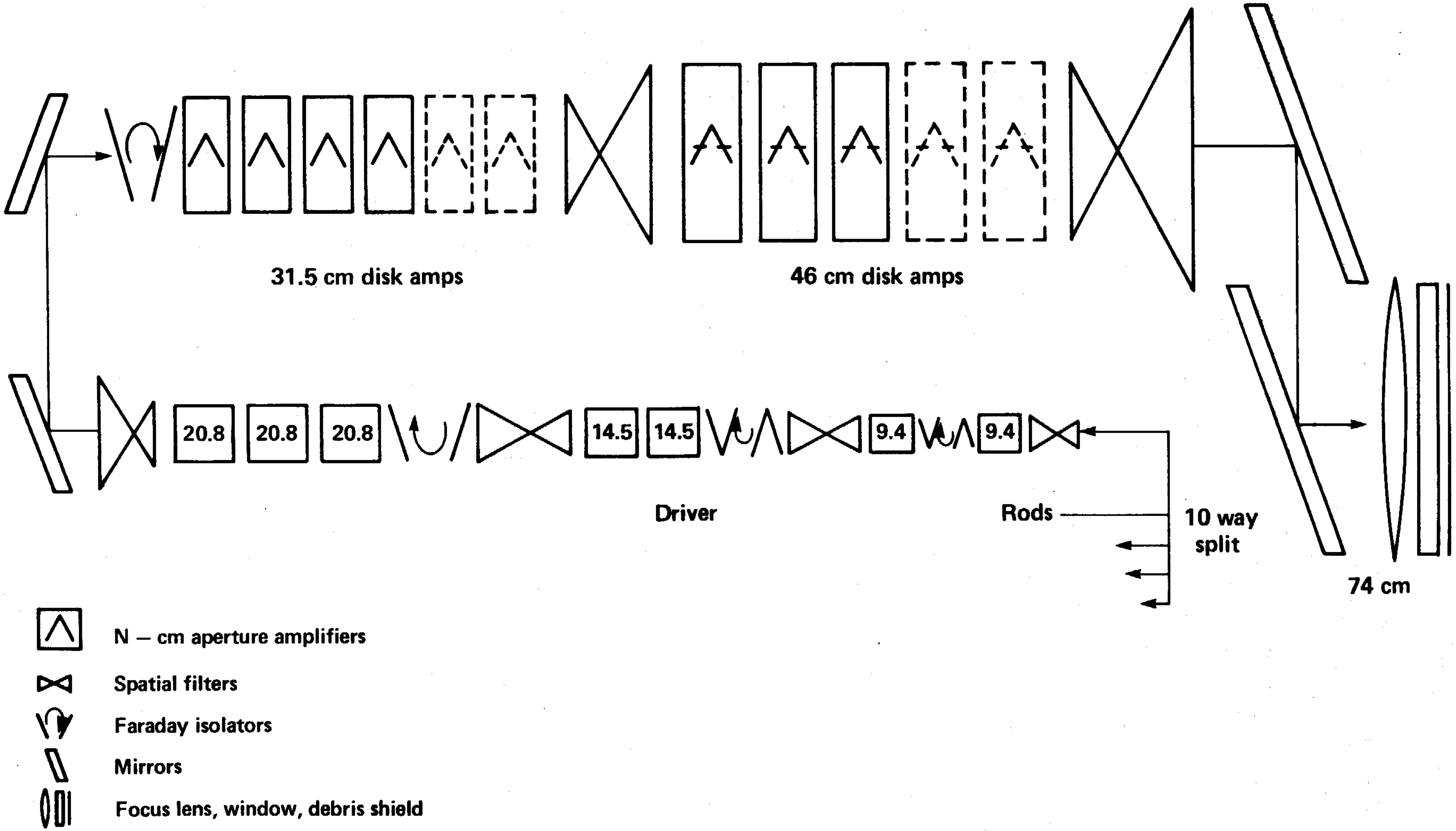

31.5 cm disk amps
46 cm disk amps
20.8
20.8
20.8
14.5
14.5
9.4
9.4
Driver
Rods
10 way split
74 cm
N – cm aperture amplifiers
Spatial filters
Faraday isolators
Mirrors
Focus lens, window, debris shield

Figure 1

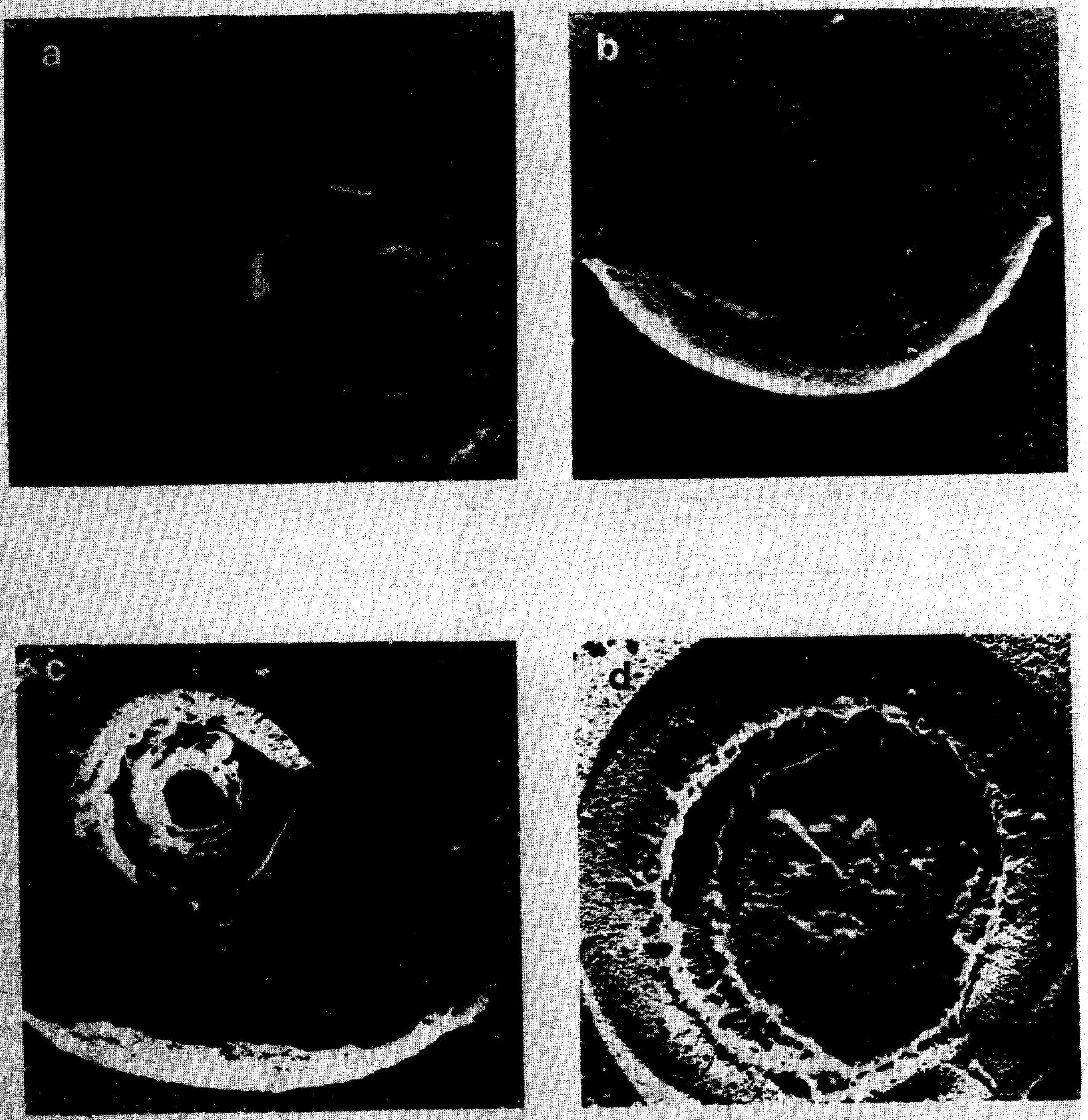
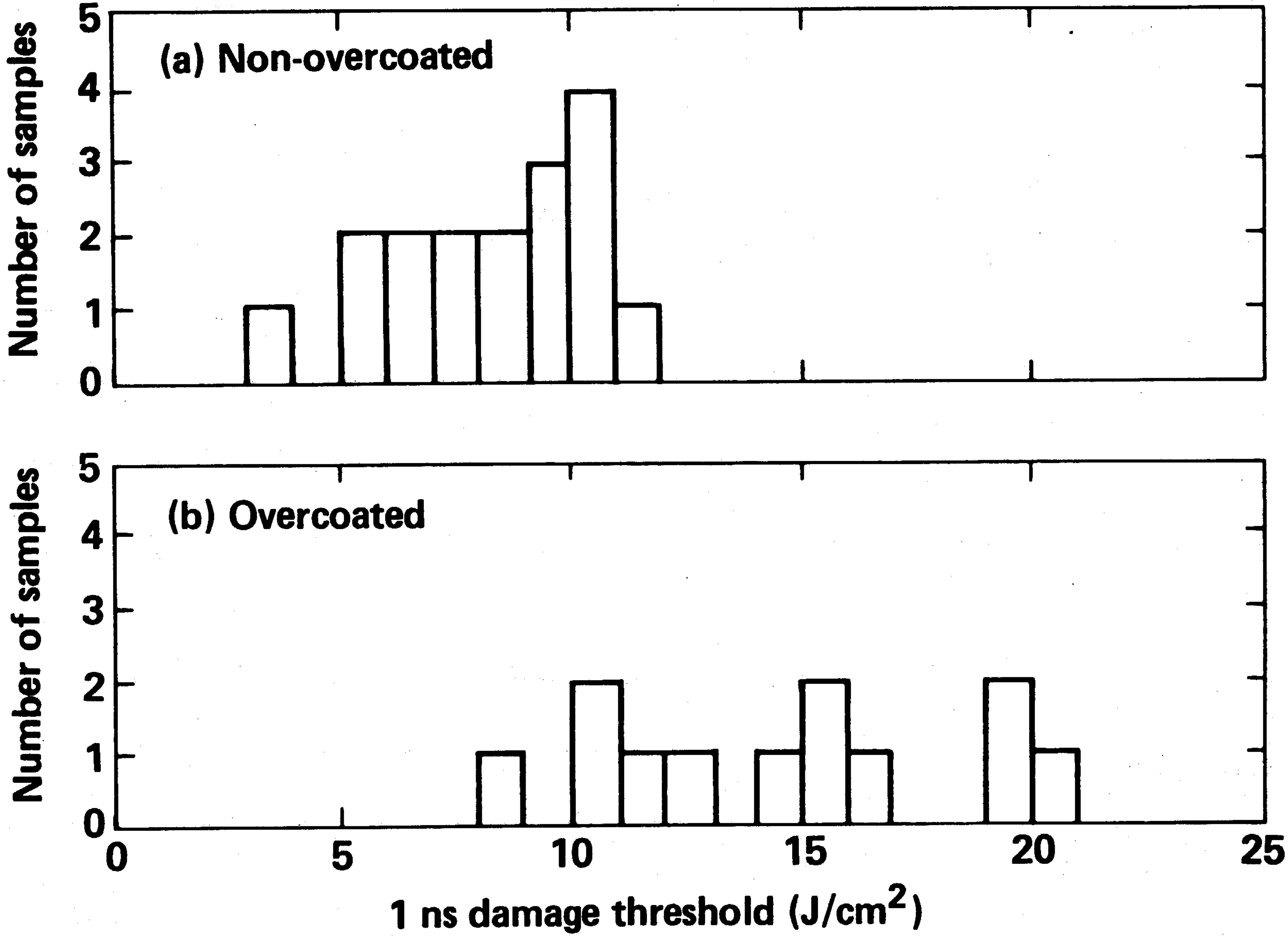



Figure 2



(a) Non-overcoated
(b) Overcoated
Number of samples
Number of samples
0
1
2
3
4
5
0
5
10
15
20
25
1 ns damage threshold (J/cm2)


Figure 3

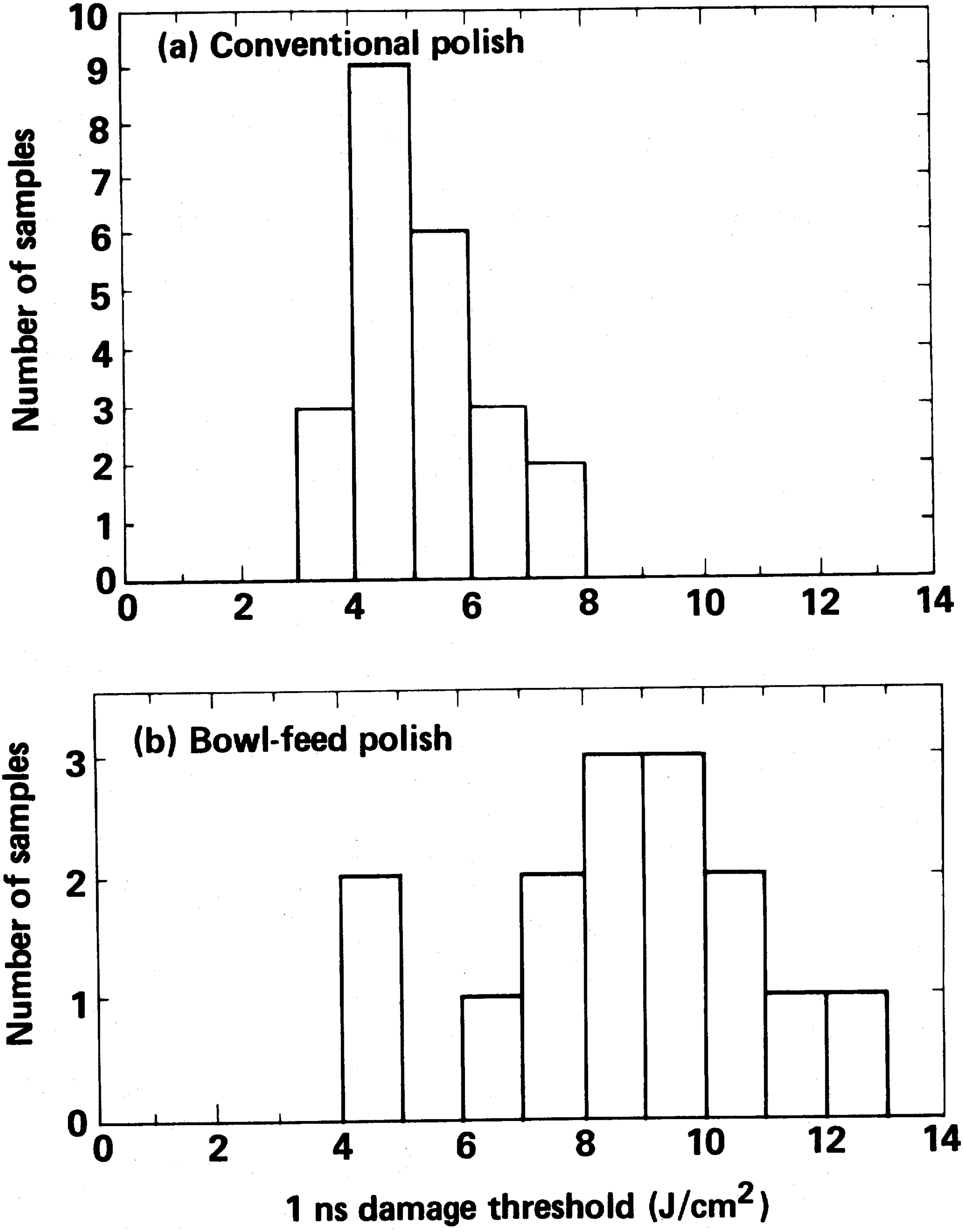

(a) Conventional polish
Number of samples
0
1
2
3
4
5
6
7
8
9
10
0
2
4
6
8
10
12
14
(b) Bowl-feed polish
Number of samples
0
1
2
3
0
2
4
6
8
10
12
14
1 ns damage threshold (J/cm2)

Figure 4